

Operando Soft X-ray Absorption Studies of Charge Compensation in Li-ion Cathode Materials

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One of the persistent puzzles in Li battery research is the cooperative roles of oxygen and transition metal species in the charge-compensation reactions during battery operation. The instability of transition metal oxide cathodes, to the point of oxygen evolution in some cases, is well-known. While hard X-ray XANES on these materials reveal that although the charge compensation mechanism is based on TM ion oxidation, some of the reversible charge capacity cannot be explained: oxygen is suspected of oxidizing to compensate charge lost to delithiation. In order to probe the oxygen 2p and TM 3d orbitals in these oxides without interference from liquid electrolytes, we have developed a new battery Georgia Tech using combines a glass ceramic electrolyte. The absence of a liquid electrolyte in this novel cell design allows operando studies of the cathode from the cathode-vacuum interface to the interior bulk cathode during delithiation using near-surface and bulk-sensitive signals, respectively. The LiMO₂ cathodes, oxidation of oxygen is revealed to proceed in parallel with TM oxidation, thereby explaining the fundamental origins of oxygen evolution. Applications of this operando cell to further studies of gas exposure and gas evolution experiments will be discussed.